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I am knowledgeable in the English language, and in the German language in which the below identified application was filed, and that I believe the annexed English translation of the certified copy of German Patent Application 101 23 604.2 to be a true and complete translation of the above identified application.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardise the validity of the application or any patent issued thereon.

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Priority certificate relating to the filing of a patent application

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Applicant/Proprietor:	Solvay Fluor und Derivate GmbH, Hannover/DE
Title:	Non-combustible polyester polyol and/or polyether polyol premix for the production of foamed products
IPC:	C 08 G, C 08 K, C 08 J

The appended items are a true and exact reproduction of the original documents of this patent application.

Munich, 21st February 2002
German Patent and Trade Mark Office
The President

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(signature)
Waasmaier

Abstract

The invention relates to non-combustible polyether polyol and/or polyester polyol premixes for the production of foamed products, in particular PU foams, the premix consisting of polyol, more than 4% by weight of a binary blowing-agent mixture consisting of HFC 365mfc and a further fluorohydrocarbon and 10 to 20% by weight of a phosphorus-based flameproofing agent.

Claims

1. A non-combustible premix for the production of foamed products from polyether polyol and/or polyester polyol, additions such as catalyst, stabiliser, further additives and blowing agent, characterised by 4 to 35% by weight of a binary blowing-agent mixture and 10 to 20% by weight of a phosphorus compound.

2. A non-combustible premix according to Claim 1, characterised in that a mixture of a) 1,1,1,3,3-pentafluorobutane and b) 1,1,1,2-tetrafluoroethane, 1,1,1,2,3,3,3-heptafluoropropane or 1,1,1,3,3-pentafluoropropane is contained therein as binary blowing-agent mixture.

3. A non-combustible premix according to Claim 1 and 2, characterised in that it contains 10 to 15% by weight binary blowing-agent mixture.

4. A non-combustible premix according to Claims 1 to 3, characterised in that triethyl phosphate or tris-chloroisopropyl phosphate is contained therein as phosphorus compound.

5. A non-combustible premix according to Claims 1 to 4, characterised in that it contains 10 to 15% by weight of the phosphorus compound.

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Non-combustible polyester polyol and/or polyether polyol premix for the production of foamed products

Description

The present invention relates to non-combustible polyester polyol and/or polyether polyol premixes for the production of foams, in particular of polyurethane foam products.

Polyurethane foams are produced by reaction of isocyanates with a polyol or a polyol mixture in the presence of blowing agents, preferably hydrofluoroalkanes.

The use of 1,1,1,3,3-pentafluorobutane (HFC365mfc) as a blowing agent for the production of polyurethane foams is known. Since 1,1,1,3,3-pentafluorobutane has a flashpoint of below -27°C, it is regarded as a readily flammable liquid and limits are set on its use as a blowing agent. Usually, therefore, 1,1,1,3,3-pentafluorobutane is used in a mixture with other fluorohydrocarbons.

Known blowing-agent mixtures contain in addition to HFC365mfc e.g. 1,1,1,2-tetrafluoroethane (HFC-134a) or 1,1,1,2,3,3,3-heptafluoropropane (HFC 227ea) or 1,1,1,3,3-pentafluoropropane (HFC 245fa). These blowing-agent mixtures have no flashpoint and are suitable for the production of foamed plastics.

It is likewise known and conventional, in order to produce foams, first to produce what are called premixes from the different feed materials, and these are then reacted with

the isocyanate. For the preparation of the premix, polyols or polyethers, blowing agents, catalysts and optionally further additives are mixed together in the required amounts. The foams are then produced by bringing the premix into contact with the isocyanate or isocyanates.

If premixes are prepared using the aforementioned blowing-agent mixtures, when a critical quantity of blowing agent is exceeded surprisingly it may happen that the entire system has to be classified as combustible owing to the low flashpoint, although the blowing-agent mixture and polyol system per se are not combustible.

The object of the invention is to provide a non-combustible, stable premix for the production of foams which do not have a flashpoint, even with a blowing-agent content of more than 4% by weight in the system.

Premixes according to the invention consist of

a) polyol: preferably polyether polyols or polyester polyols are used.

b) 4 to 35% by weight, preferably 10 to 15% by weight, blowing-agent mixture, the blowing-agent mixture containing in addition to HFC365mfc at least 5% by weight, preferably 7% by weight, of a further fluorohydrocarbon, preferably HFC134a, HFC227ea or HFC245fa, and

c) 10 to 20% by weight, preferably 10 to 15 % by weight, of a phosphorus compound, preferably triethyl phosphate or tris-chloroisopropyl phosphate.

Known phosphorus-based flameproofing agents may likewise be used as phosphorus compound.

Further additions, such as catalyst, stabiliser and further additives are admixed to the premix in known manner.

The premix according to the invention is contacted with the isocyanate or isocyanates and foamed in known manner.

Usually polyisocyanates for example with 2 to 4 isocyanate groups are used for the production of the polyurethane foams. Their preparation and the basic chemicals usable therefor are known.

These isocyanates have an aliphatic hydrocarbon radical with up to 18 C atoms, a cycloaliphatic hydrocarbon radical with up to 15 C atoms, an aromatic hydrocarbon radical with 6 to 15 C atoms or an araliphatic hydrocarbon radical with 8 to 15 C atoms. Starting constituents which are particularly preferred industrially are for example 2,4- and 2,6-toluylene diisocyanate, diphenylmethane diisocyanate, polymethylene polyphenyl isocyanate and mixtures thereof. Also what are called modified polyisocyanates, which contain carbodiimide groups, urethane groups, allophanate groups, isocyanurate groups, urea groups or biuret groups, may be used.

Further starting constituents are compounds with at least 2 hydrogen atoms which are reactive with respect to isocyanates. These are in particular compounds with a molecular weight of 400 to 10,000, which preferably contain 2 to 8 hydroxyl groups and furthermore may contain amino groups, thiol groups or carboxyl groups.

Additionally chemical blowing agents such as water can be added as further auxiliaries and additives to the system which is to be foamed. Catalysts such as for example tertiary amines, such as dimethylcyclohexylamine, and/or organic metal compounds can also be used. Surface-active additions such as emulsifiers or foam stabilisers, for example siloxane polyether copolymers, reaction-delaying agents, cell

regulators such as paraffins, fatty alcohols or dimethylpolysiloxanes, pigments and dyes, may be used. Furthermore, stabilisers against the effects of ageing and the weather, fillers, dyes, antistatic agents, nucleating agents, pore-regulating substances or biocidal active substances may be used.

Suitable catalysts are for example mentioned in international patent application WO 96/14354. These include organic amines, aminoalcohols and aminoethers such as morpholine compounds, for example dimethylcyclohexylamine, diethanolamine, 2-dimethylaminoethyl-3-dimethylamino-propylether, 2-dimethylaminoethylether, 2,2-dimorpholino-diethylether, N,N-dimethylaminoethylmorpholine and N-dimethylmorpholine. Also organometallic compounds such as for example tin, cobalt or iron compounds can be used as catalyst. Examples which can be used are tin dioctoate, cobalt naphthenate, dibutyltin dilaurate and iron acetonylacetate.

The advantage of the premix according to the invention is that obviously the solubility characteristics of the constituents are modified due to the addition of flameproofing agents such as triethyl phosphate, tris-chloroisopropyl phosphate and further phosphates or phosphonates, so that the flashpoint rises and the classification "combustible" no longer applies. Thus simple storage and transport of the premix is possible.

Example 1:

Polyether polyol (Tercarol A350) was mixed with 10% by weight blowing agent (relative to polyol) and the flashpoint was determined. A binary mixture of 94% HFC 365mfc and 6% by weight HFC 227ea was used as blowing agent.
Flashpoint according to DIN EN ISO 13736: 15°C

Examples 2 to 5:

A premix was prepared analogously to Example 1 from polyether polyol (Tercarol A350) and 10% by weight binary blowing-agent mixture and also additionally triethyl phosphate (TEP) or tris-chloroisopropyl phosphate (TCPP) were added. The flashpoint was determined in accordance with DIN EN ISO 13763. No flashpoint could be determined.

Table 1

Examples	Blowing-agent mixture	Ratio	Phosphorus compound
2	HFC 365mfc/HFC 227ea	94:6	10% by weight TEP
3	HFC 365mfc/HFC 227ea	94:6	13% by weight TCCP
4	HFC 365mfc/HFC 134ea	93:7	15% by weight TEP
5	HFC 365mfc/HFC 245ea	75:25	10% by weight TEP